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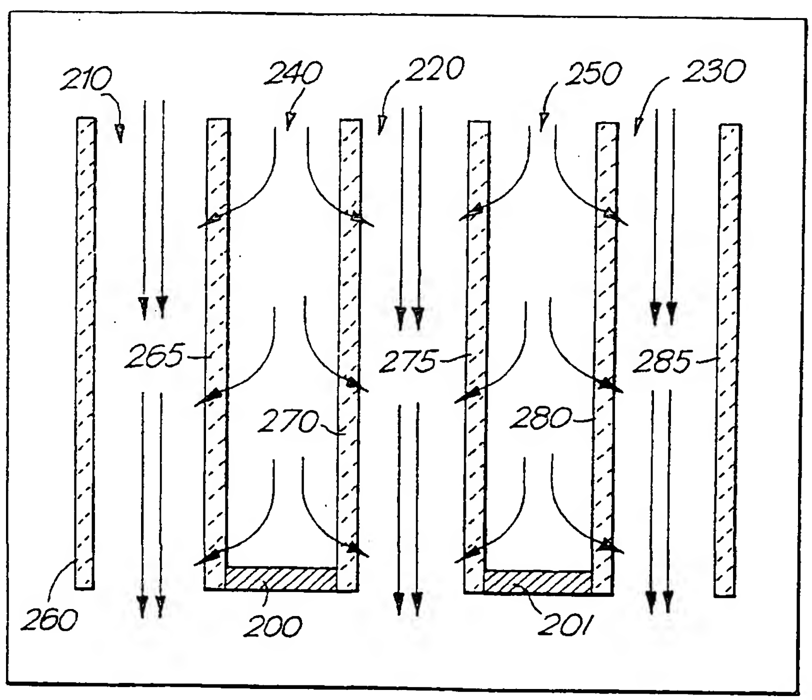
(54) Title: MONOLITHIC CATALYST/FILTER APPARATUS

(57) Abstract

An apparatus for the control of gaseous and particulate emissions from a gaseous stream containing particulate material, or a gaseous stream without particulate material. The apparatus comprises an inlet and an outlet, and a plurality of cells having walls (260, 265, 270, 275, 280, 285) made of a material permeable to the gaseous stream. The cells are oriented to allow flow (210, 220, 230, 240) of the gaseous stream from the inlet to the outlet, and each of the cells has a first end which is adjacent the inlet of the apparatus, and a second end which is adjacent the outlet of the apparatus. The walls are coated with a catalyst. A barrier (200, 201) is provided at some, but not all, of the second ends of the cells, the barrier comprised of a material that does not allow passage of the gaseous stream.



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MONOLITHIC CATALYST/FILTER APPARATUS

This invention relates generally to monolithic catalysts. More particularly, this invention relates to monolithic catalysts which perform both filtering and catalysis functions to treat gaseous streams containing particulate material.

Government emissions standards, as well as a desire to prevent environmental pollution, have led to the development of many technologies which promote the oxidation of unburned hydrocarbons and the removal of other deleterious species found in the exhaust gases of internal combustion engines. These unburned hydrocarbons, along with carbon monoxide and nitrogen oxide are often dealt with in catalytic converters that are placed in the exhaust gas line. Typical catalytic beds might comprise one or more platinum group metals supported on alumina. Other catalytic materials are, of course, known.

Other pollutants are also common in combustion exhaust lines, especially diesel exhaust lines. In addition to those pollutants mentioned above, soot-containing particulate material is produced during combustion. Such particulate materials are also deleterious to the environment, and the removal of these components prior to discharging the exhaust stream to the atmosphere is desirable.

Soot is essentially carbonaceous material and, where it is generated during combustion of diesel fuel in the cylinder of diesel engine, leaves the engine with a layer of soluble organic fraction (SOF) surrounding it. That soot, covered in the SOF is one kind of particulate material that can be problematic.

Technologies have been developed to counter the particulate emission. One example includes the subject of United States Patent No. 4,902,487 to Cooper et al. That technology involves passing diesel exhaust gas through a filter to remove particulate material wherein the particulate material is combusted in the presence of NO_2. The NO_2 may be obtained through oxidation of NO upstream of the filter. Such apparatus can achieve a very high conversion of the diesel exhaust particulates. A possible problem with such a system exists

however, if the engine malfunctions and creates excessive particulate material (i.e. particulate amounts in excess of that produced during normal engine operation).

The excess particulate material can block the filter. Blockage of the filter begins with an increase in pressure drop across the filter and ends with an insurmountable blockage that can damage the engine and other parts of the system.

An alternative method of treating an exhaust stream containing particulate material is to use only an oxidation catalyst. In such a case, relatively low conversions of SOF are achieved. Because no filter is used, however, blockage and its associated problems (as discussed above related to the use of a filter) do not normally occur.

An improved filter/catalyst apparatus would provide effective filtering and catalysts while preventing the harmful effects on the system associated with blockage by particulate material which may be present in the exhaust stream.

The present invention provides an apparatus for the control of gaseous and particulate emissions from a gaseous stream containing particulate material with carbonaceous combustible content. The invention is particularly applicable to diesel exhaust streams, but applies to any stream that contains noxious or otherwise undesirable gases. The invention is particularly applicable to streams containing both noxious or otherwise undesirable gases and particulate material. The apparatus comprises an inlet side and an outlet side and a plurality of passageways, or cells. The interior walls which separate the cells are made of a material which is permeable to the gaseous stream. The cells are oriented to allow flow of the gaseous stream from the inlet to the outlet of the apparatus, and each of the cells has a first end adjacent the inlet of the apparatus and a second end adjacent the outlet of the apparatus. The cell walls are coated with a catalyst. A barrier is present in some, but not all, of the second ends of the cells. The barrier may be comprised of a material that does not allow passage of the gaseous stream or permits passage of gases but not particulate. Thus, the cells, as a group, are partially blocked such that the exhaust stream can pass freely through some of the cells but not all of the cells.

The invention is best understood from the following detailed description when read in connection with the accompanying drawings. It is emphasized that, according to common practice, various features of the drawings are not to scale. On the contrary, the dimensions of the various features are arbitrarily expanded or reduced for clarity. The drawings include the following figures:

Fig.1 shows a schematic view of an apparatus according to the present invention;
Fig.2 shows a schematic view of a cross section of an apparatus according to the present invention;
Fig.3 is a plot showing the opacity reduction where only a monolithic catalyst is used; and
Fig.4 is a plot showing the opacity reduction where the apparatus of the present invention is used.

The present invention provides an apparatus for the control of gaseous and particulate emissions from a gaseous stream containing particulate material with carbonaceous combustible content. The invention is particularly applicable to diesel exhaust streams. The apparatus comprises an inlet side and an outlet side and a plurality of cells. The cell walls are made of a material that is permeable by the gaseous stream but largely not permeable by the particulate material.

Fig.1 shows a schematic view of an apparatus according to the present invention. Cells 100 are contained in a region of the apparatus which has an inlet and an outlet, shown by arrows which indicate gas flow. As the exhaust gas enters the apparatus, it passes through the cells and eventually exits through the other side where it is vented to atmosphere, or further treated. A typical apparatus is placed directly into the exhaust line. The size of a typical apparatus might be about 15cm (6 inches) long by about 20-25cm (8 – 10 inches) in diameter, for a vehicle using a 6 liter diesel engine.

Fig. 2 shows a schematic view of a cross section of a portion of an apparatus according to the present invention. The cells are oriented to allow flow of the gaseous stream from the inlet to the outlet of the apparatus, and each of the cells has a first end

adjacent the inlet of the apparatus and a second end adjacent the outlet of the apparatus. The cell walls are coated with, or otherwise contain, a catalyst. A barrier is present in some, but not all, of the second ends of the cells. The barrier is comprised of a material that does not allow passage of the gaseous stream. Thus, the cells, as a group, are partially blocked such that the exhaust stream can pass freely through some of them but not all of them. In the preferred embodiment, the cells are alternatively blocked. In other words, the cells are periodically blocked – i.e. one blocked, one open, one blocked, etc., as shown schematically in both Figs. 1 and 2.

In Fig. 2, the arrows indicate gaseous flow of an exhaust stream through the cells. Barriers 200 and 201 are present in alternating cells. In the case illustrated, the cells are straight cells, but the cells do not have to be straight. Open cells 210, 220, and 230 allow easy passage of the gaseous stream. Closed cells 240 and 250 do not allow easy flow, as barriers 200 and 201 are present at the end of the cells.

Walls 260, 265, 270, 275, 280, and 285 are all permeable to the gaseous stream, but are substantially not permeable to the particulate material that is present in the stream. Cells 210, 220, and 230 all allow the gaseous stream including particulate to pass freely through, as indicated by the arrows in Fig. 2. Cells 240 and 250 do not allow such free flow, however, due to barriers 200 and 201. In cells 240 and 250, gas is forced through the semi permeable walls and into cells 210, 220, and 230, as indicated by the arrows in Fig. 2. The walls are considered semipermeable because they do not allow the passage of most of the particulate material (some particulate material may work its way through, but the vast majority is captured at the surface of the wall).

Thus, as the gaseous stream passes through the walls, the particulate material is collected on the surface of wall 265 which is exposed to cell 240, the surface of wall 270 which is exposed to cell 240, the surface of wall 275 which is exposed to cell 250, and the surface of wall 280 which is exposed to cell 250.

Gas is forced through the walls from the blocked cells to the open cells because of pressure generated in the blocked cells. But additional effects of the physical arrangement are realized. One advantage is that the open cells act as eductors. The decrease in pressure at the surface of walls exposed to the open cells as gas flows through them increases the pressure drop across the wall and effectively draws the gas through the wall.

As particulate material accumulates on the walls, it is typically catalysed and combusted along, or on, the coated wall. Uncombusted, inorganic ash or other uncombustible material will fall free and out of the system due to forced gas flow, its own weight, and/or vibrations caused from the engine operation and other movement of the housing. Even if the particulate material does not fall free, however, blockage of the apparatus will never occur from particulate accumulation, however, because some of the cells are open to flow. Similarly, in the case of an engine malfunction such that an excess of particulate material enters the apparatus, no blockage will occur. Thus, a compromising situation exists such that some particulate material is collected and some emission components are catalytically reacted to form less harmful components, but the system will never block and cause engine damage. The overall effect is to reduce harmful particulate and gaseous emissions while preventing blockage which would result in engine damage.

An additional, somewhat related, advantage is realized when the gas flowing through the open cells demonstrates a laminar flow profile. Where the gas flow is laminar, there is, relative to turbulent flow, reduced contact between the gas stream and the catalyst on the surface of the walls. By utilizing the apparatus according to the present invention, the wall-flow eduction from the closed cells to the open cells causes a disruption in the laminar flow though the open cells which enhances mass transfer to the catalytic surface and increases catalytic activity.

The amount of cells in a given apparatus can vary depending on the particular application. For example, for an average 6 liter diesel engine, about 15.5 cells per sq cm (100 cells per square inch) of cross section (cpsi) would probably suffice. Of course, specific operating conditions and environment would impact this design parameter.

For example, differing engine designs, exhaust line designs and even fuels would require, possibly, different cell sizes. Apparati according to the present invention could have cell sizes up to about 31 cells per sq cm (200 cpsi), and even higher, depending on the necessary wall thicknesses. Other than design choice, manufacturing constraints can also create limits on cell concentration. Conveniently, half the cells are provided with the barrier.

The walls can be constructed from any suitable material which meets the needs described above. For example, the walls may be made of a woven or knitted wire mesh or perforated metal or a suitable ceramic material. A preferred wall material is cordierite (a crystalline mineral comprising a silicate of aluminum and magnesium), but silicon carbide and stainless steel mesh may also be used. The walls can be suitably designed for each engine application as the particulate emissions differ from engine to engine.

The catalyst coating on the walls may also be of any suitable catalyst for the desired purpose. It may, for example, be any that is effective to convert NO to NO_2 (NO_2 is an effective oxidizer, and its presence is therefore desirable for subsequent oxidation of particulate material). It may also be a high platinum loading catalyst carried on a ceramic or metal honeycomb catalyst support, separate from the wall material itself. Alternatively, the wall itself may be the support. The catalyst layer may also be an alumina-based washcoat impregnated with platinum group metals (i.e. platinum, palladium, ruthenium, rhodium, osmium, iridium, etc.). The catalyst may be a base metal, with or without any platinum group metal. The base metal may be by itself, as in a base metal washcoat, or be present in conjunction with alumina. Typical base metals include manganese, copper, iron, nickel, cobalt, and oxides of these metals. In addition, rare earth elements could comprise the base metal. Such rare earth base metals would include cerium, lanthanum, praseodymium, neodymium, etc. (atomic numbers 57-71) and oxides of these elements. Of course, combinations of these metals and their oxides could also be used. Many other catalyst and catalyst configurations can be employed in conjunction with the present invention and one skilled in the art can make the selection based upon each situation and the desired catalytic activity.

When the catalyst is applied to the wall, care is taken not to block the pores which allow flow of the gaseous stream through the walls. Through appropriate methods of applying the catalyst, known to those skilled in the art, this blockage problem may be avoided.

Figs. 3 and 4 show results for opacity testing of exhaust streams from different treatment apparati. Opacity is defined as the fraction of light transmitted from a source which is prevent from reaching the observer or instrument receiver, expressed in percentage. Thus, something which has opacity of 100% is completely opaque, and something with 0% opacity is completely transparent. For the purpose of generating the comparative data shown in Figs. 3 and 4, opacity measurements were taken across the exhaust stream as it traversed the test chamber. In Fig. 3, this reading was recorded as the exhaust stream exited only a monolithic catalyst chamber. In Fig. 4, the reading was recorded as the exhaust stream exited an apparatus according to the present invention.

Both Figs. 3 and 4 show the percent decrease in opacity (i.e. the percent change in percentage of light transmitted through the exhaust stream as it exited the apparatus as compared to before it entered the apparatus). Thus, a greater percent decrease in opacity translates to a greater removal of particulate material. Fig. 3 shows the opacity reduction where only a monolithic catalyst is used, without the alternating blocked/unblocked cells according to the present invention. Data was generated using a 6 liter diesel engine and was collected over an approximate 5 month period. Measurements were taken four times on each day indicated and averaged. In each figure, two sets of data as shown for each measurement period. One set was taken from exhaust exiting the system while the engine was operating and the rpm was ramped from 750 to 2300 rpm within four seconds. Similarly, a second set of data was taken from exhaust exiting the system while the engine was ramped from 1500 to 2300 rpm in four seconds. As can be seen in Fig. 3, the decrease in opacity reduction where all cells are open is between approximately 10 and 20% in most cases.

Fig. 4 shows the opacity reduction where the apparatus is configured according to the present invention, that is, as shown schematically in Fig. 1. Here, opacity reduction

ranges, approximately, between 30 and 50%. As in Fig. 3, data was collected from the use of a 6 liter diesel engine over the same approximate 5 month period. Two sets of data are shown for each measurement period. One set was taken from exhaust exiting the system while the engine was operating and was ramped from 750 to 2300 rpm in four seconds. Similarly, a second set of data was taken from exhaust exiting the system while the engine was operating and was ramped from 1500 to 2300 rpm in four seconds. Again, four data points were generated for each day under each operating condition and were then averaged to generate the point on the graph.

As can be seen through consideration of the data represented, where the apparatus of the present invention is utilized, there is an increase in the reduction of opacity. Thus, by utilizing the apparatus of the present invention, overall removal of particulate material was increased as compared to the use of only a monolithic catalyst. The present invention provides a system where particulate removal is enhanced and necessary catalytic activity is promoted, while simultaneously removing the possibility of the system becoming blocked (and subsequently damaged) due to excessive particulate material depositing as may arise from engine malfunction.

As discussed above, even where no particulate material is present in the gaseous stream, the present invention provides an advantageous system in which catalytic activity is enhanced. By blocking some channels and allowing free flow through others, the enhanced turbulence created by wall flow eduction increases catalyst efficacy. Through enhanced mass transfer within the system, overall oxidation and related combustion processes and chemical reactions are improved.

Although illustrated and described herein with reference to certain specific embodiments, the present invention is nevertheless not intended to be limited to the details shown. Rather, various modifications may be made in the details within the scope and range of equivalents of the claims and without departing from the spirit of the invention.

CLAIMS

1. An apparatus for the control of gaseous and particulate emissions from a gaseous stream containing particulate material, said apparatus comprising: an inlet and an outlet; a plurality of cells having walls made of a material permeable to said gaseous stream, said cells oriented to allow flow of said gaseous stream from said inlet to said outlet, each of said cells having a first end adjacent said inlet of said apparatus, and a second end adjacent said outlet of said apparatus, said walls coated with a catalyst; characterised in that some, but not all, of said second ends of said cells are provided with a barrier, said barrier comprised of a material that either does not allow passage of said gaseous stream or permits passage of gases but not of particulate, and that the remaining cells provide for unrestricted flow of said gaseous stream.

2. The apparatus of claim 1, wherein every other of said cells has a said barrier.

3. The apparatus of claim 1, or 2 wherein there are up to about 31 cells per sq cm (200 cells per square inch) of said apparatus.

4. The apparatus of claim 1, 2 or 3 wherein said catalyst is comprised of a metal oxide based washcoat impregnated with one or more metals selected from the group consisting of platinum, palladium, ruthenium, rhodium, osmium, iridium, and combinations thereof.

5. The apparatus of any one of the preceding claims wherein said catalyst comprises a base metal.

6. The apparatus of any one of the preceding claims wherein said walls are comprised of cordierite.

Fig.1.

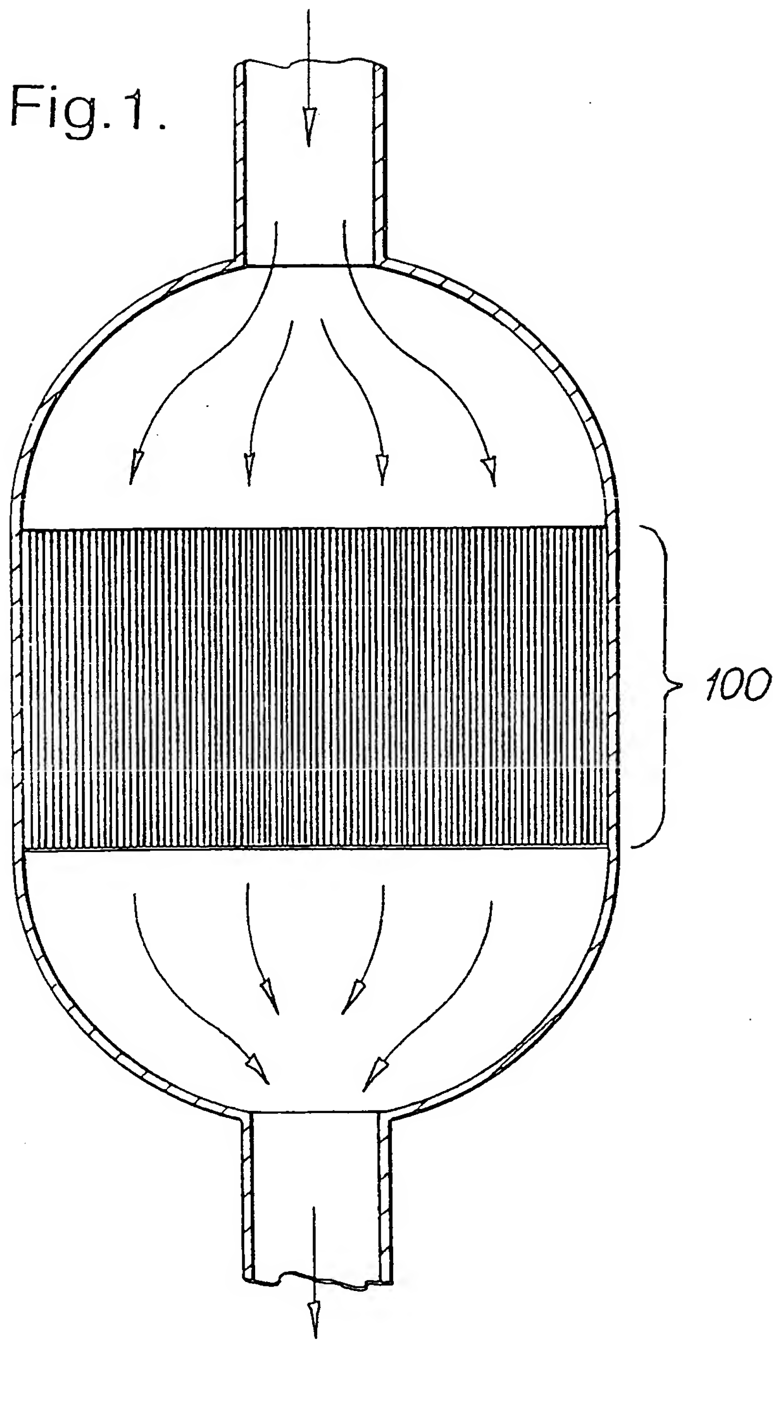

Fig.2.

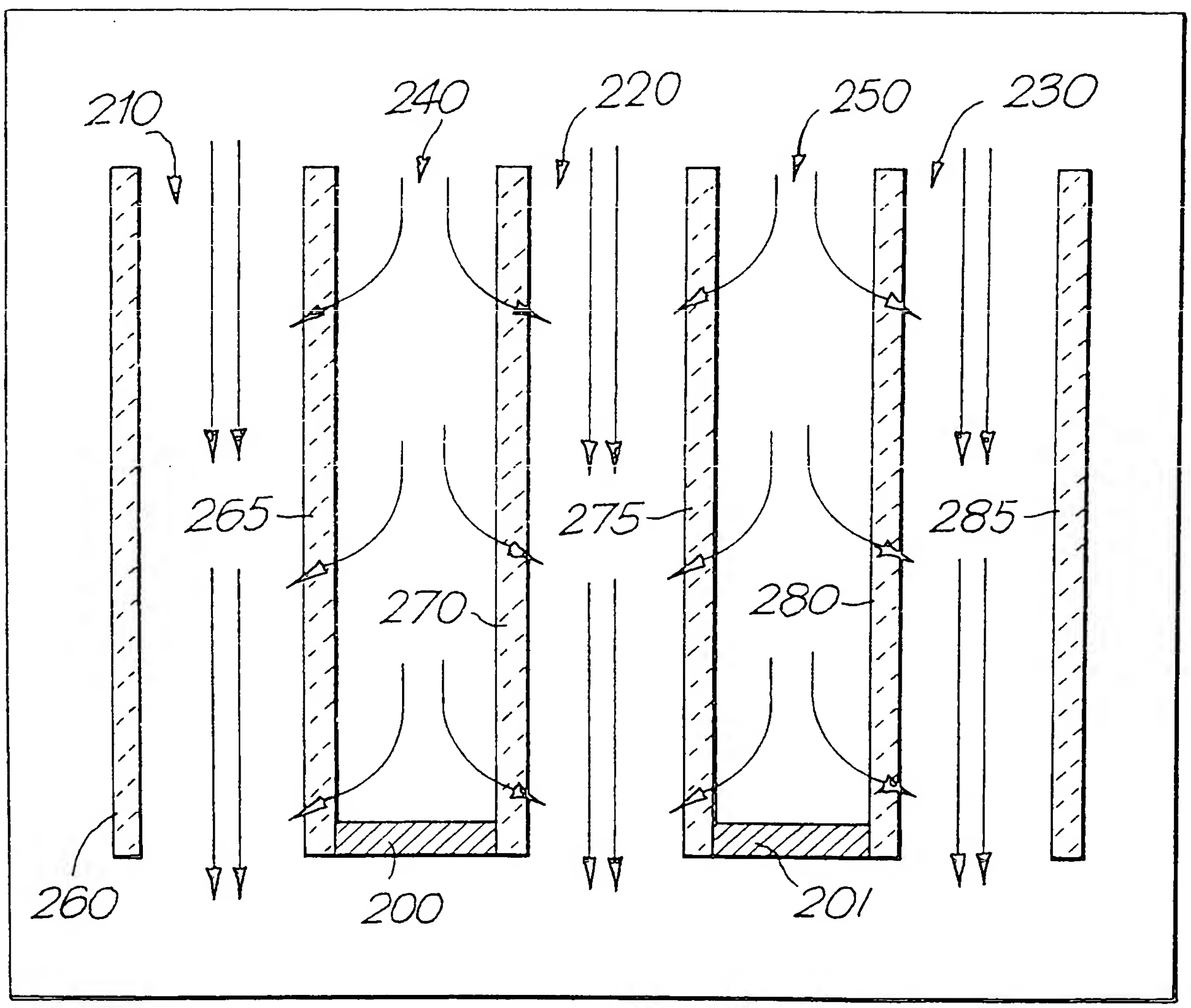

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Fig.3.

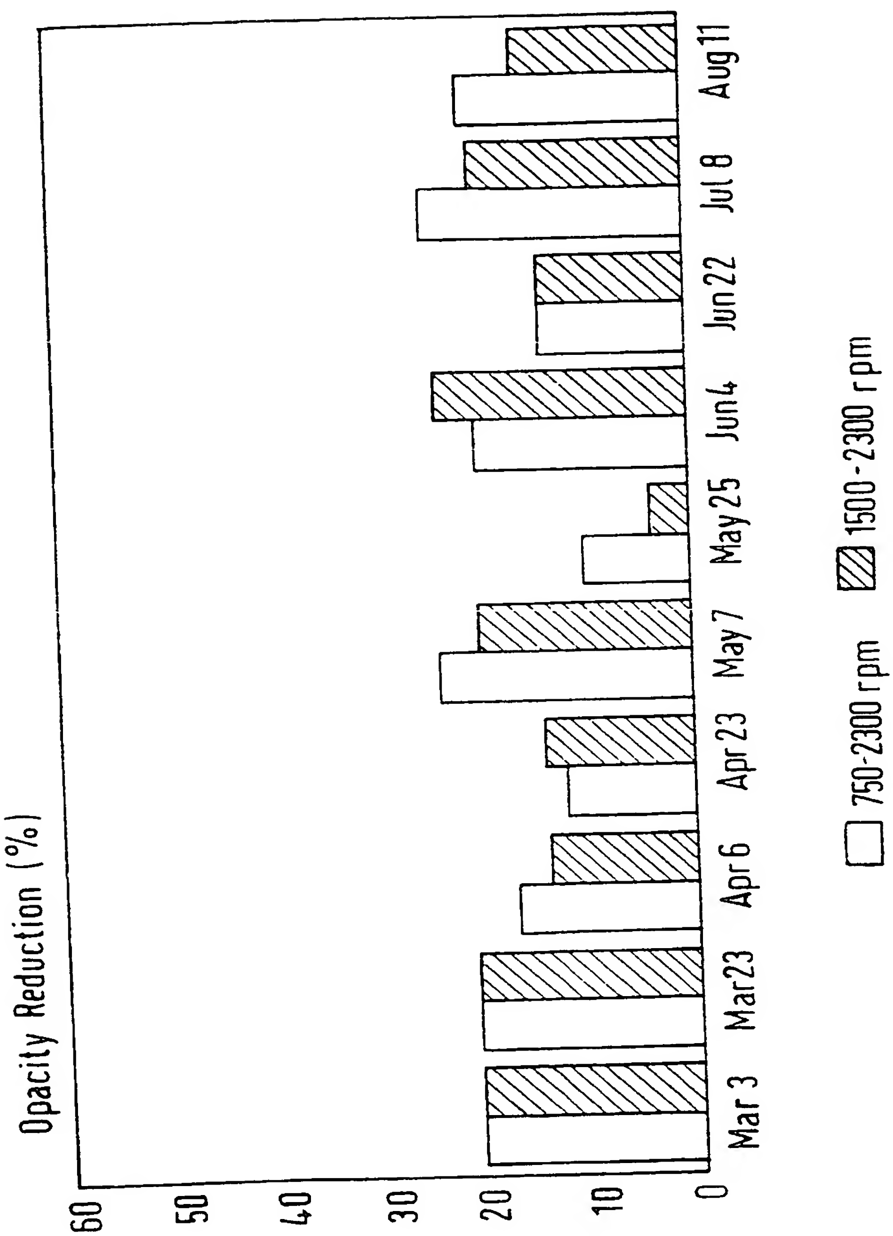
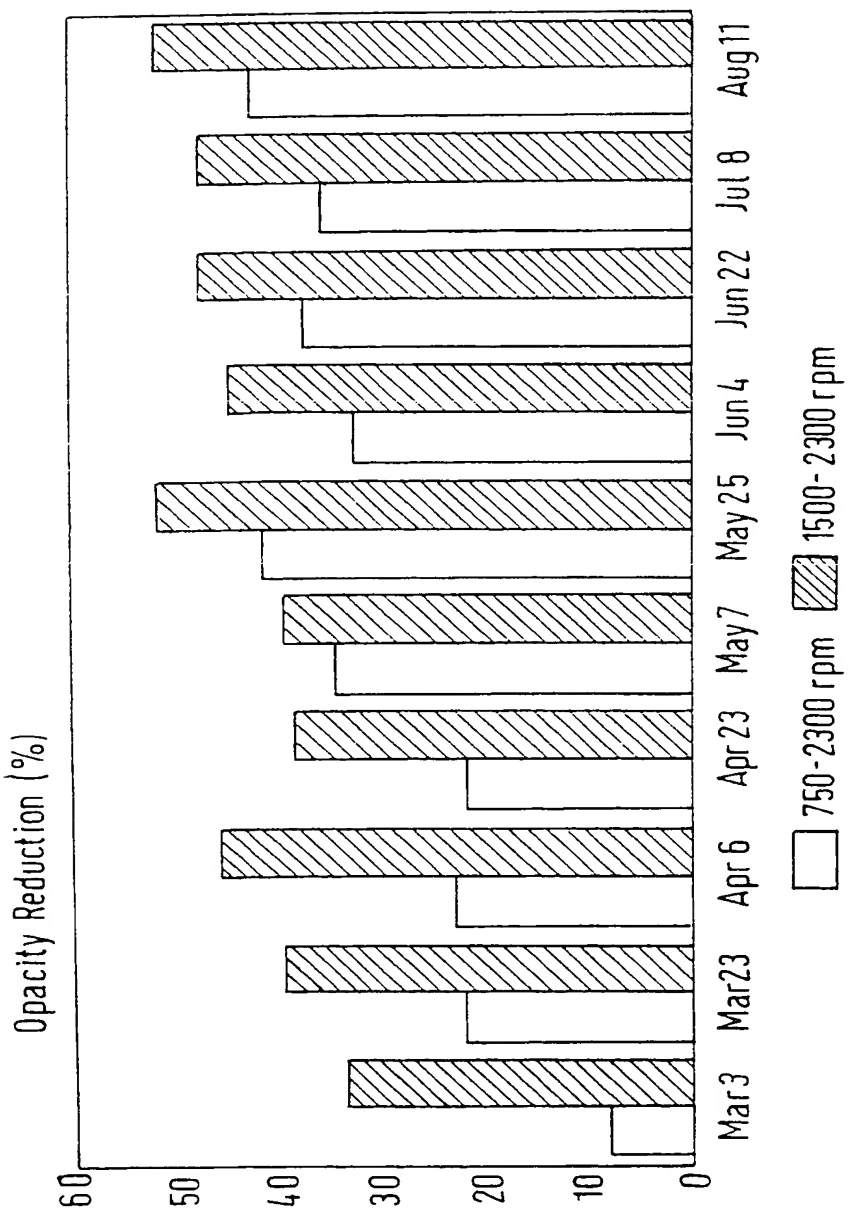

Opacity Reduction (%)
60
50
40
30
20
10
0
Mar 3
Mar23
Apr 6
Apr 23
May 7
May 25
Jun 4
Jun 22
Jul 8
Aug 11
750-2300 rpm
1500-2300 rpm

Fig.4.


Opacity Reduction (%)
60
50
40
30
20
10
0
Mar3
Mar23
Apr6
Apr23
May7
May25
Jun4
Jun22
Jul8
Aug11
750-2300 rpm
1500-2300 rpm

INTERNATIONAL SEARCH REPORT

International Application No
PCT/GB 00/00435

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 F01N3/035 B01J35/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 F01N B01D B01J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

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C. DOCUMENTS CONSIDERED TO BE RELEVANT

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
7 April 2000	14/04/2000

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International Application No
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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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